

Quasi-linear magnetoresistance and the violation of Kohler's rule in the quasi-one-dimensional $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$ superconductor

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Abstract. We report on the quasi-linear in field intrachain magnetoresistance in the normal state of a quasi-one-dimensional superconductor $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$ ($T_c \sim 4.6$ K). Both the longitudinal and transverse in-chain magnetoresistance shows a power-law dependence, $\Delta\rho \propto B^\alpha$, with the exponent α close to 1 over a wide temperature and field range. The magnetoresistance shows no sign of saturation up to 50 tesla studied. The linear magnetoresistance observed in $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$ is found to be overall inconsistent with the interpretations based on the Dirac fermions in the quantum limit, charge conductivity fluctuations as well as quantum electron-electron interference. Moreover, it is observed that the Kohler's rule, regardless of the field orientations, is violated in its normal state. This result suggests the loss of charge carriers in the normal state of this chain-containing compound, due presumably to the charge-density-wave fluctuations.

1. Introduction

Understanding the normal state properties of a superconductor is the key step to reveal its pairing mechanism[1]. However, the normal state of some *unconventional* superconductors, such as the high T_c cuprates, may be sometimes more extraordinary and controversial than their superconducting counterpart[2]. In the pantheon of various superconducting materials, quasi-one-dimensional (Q1D) superconductors hold a special place in the study of correlated electrons as they display many rich physical phenomena exclusively for the reduced dimensionality, *e.g.*, the field-induced spin density wave[3], a variety of angular magnetoresistance oscillations[4, 5, 6] and even spin-triplet pairing state[7]. Notably, the normal state of some Q1D superconductors, including the organic Bechgaard salts $(TMTSF)_2X$ ($X=PF_6, ClO_4$)[8, 9] and purple bronze $Li_{0.9}Mo_6O_{17}$ [10], is often regarded as the most promising candidate to realize the so-called Tomonaga-Luttinger liquid (TLL) paradigm in the bulk materials.

In standard metals, the Lorentz force caused by an applied magnetic field changes the electron trajectory and gives rise to a positive magnetoresistance (MR) which increases quadratically with the strength of the field[11]. However, there are few exceptions where the MR may grow linearly with the field. For example, in the Dirac systems, the MR acquires the linear-in-field form once all Dirac fermions are degenerate into the lowest Landau level, *i.e.*, in the quantum limit regime[12, 13, 14]. The linear MR was also observed in some ferromagnets, including ferromagnet $Fe_{1-y}Co_ySi$ crystals[15] and geometrically constrained thin films of iron, nickel and cobalt, due to quantum electron-electron ($e-e$) interference effects[16]. Besides, quasi-linear MR was also reported in non-magnetic silver chalcogenides[17]. Other mechanisms for the linear MR involve the polycrystalline materials[18] and inhomogeneous compounds with mixed components of the resistivity tensor[19]. On the other side, the MR at a certain temperature $\Delta\rho$ under a field H obeys a general function known as the Kohler's rule: $\Delta\rho/\rho_0=f(H/\rho_0)$, where ρ_0 is the zero-field resistivity[20, 21]. As a result, the plots of $\Delta\rho/\rho_0$ as a function of H/ρ_0 at distinct temperatures will collapse onto a single curve. Interestingly, this rule, although derived from the semiclassical Boltzmann theory, was found to be well obeyed in a large number of metals, including the metals with two types of carriers, the pseudogap phase of the underdoped cuprates [22] as well as some other Q1D metals[23]. The violation of such a rule is generally believed to result from the loss of carriers with temperature or from the fact that the anisotropic electron scattering rates do not have the same T scaling on different sections of the Fermi surface (FS).

A new Q1D chain-containing compound $Ta_4Pd_3Te_{16}$ has recently been reported to be superconducting below $T_c\sim 4.6$ K[24]. In its crystal structure, one dimensional $PdTe_2$ *conducting* chains are extended along the b -axis, sandwiched by $TaTe_3$ chains and Ta_2Te_4 double chains. Soon after this finding, the low temperature thermal conductivity measurements revealed the gap node in its order parameter[25], similar to the overdoped cuprate $Tl_2Ba_2CuO_{6+\delta}$. However, the scanning tunnelling microscopy (STM) points out that its gap structure is more likely anisotropic without nodes[26].

On the other hand, both the T_c -pressure diagram and STM study suggest that the system is probably in the vicinity of an ordered state, presumably a charge-density-wave (CDW) instability[25, 27]. Recent density function calculations appear to rule out a magnetic instability as the origin of this ordered state[28]. All these findings seem to suggest that our understanding of this Q1D superconductor is far from complete. The normal state properties, which would provide valuable clues to its superconducting mechanism, have hardly been studied thus far. In this context, we study the normal state transport properties of this new Q1D superconductor and uncover that its MR, unlike most of standard metals, shows quasi-linear behaviors in a broad T and field range. Additionally, the semiclassical Kohler's rule is found to be modestly violated, in *all* three field orientations studied here. The implication of our observation has been discussed.

2. Experiment

$Ta_4Pd_3Te_{16}$ single crystals were synthesized by a solid state reaction in vacuum, following the same procedures described in Ref. [24]. The as-grown crystals have a typical size of $2.5 \times 0.25 \times 0.1$ mm³, with the longest dimension parallel to the chain direction (b -axis). X-ray diffraction (XRD) and dc magnetization measurements were performed to confirm the sample quality. The MR was measured by a standard four-probe technique with the current flowing along the b -axis for different field orientations up to 9 tesla and 13 tesla in superconducting magnets, respectively, and up to 50 tesla in the pulsed magnetic field laboratory. In this study, at least four crystals from the same growth batch were measured under different magnets and field orientations.

3. Results And Discussion

The schematic view of the crystal structure projected on the ac plane is shown in Fig. 1. We define hereafter the a^* -axis as the direction in the flat Ta-Pd-Te layer orthogonal to the chains and the c^* -axis perpendicular to the a^*b plane. The representative plot of the temperature dependence of the in-chain resistivity is given in Fig. 1, with a blow-up of its low- T superconducting transition as the inset. Clearly, the midpoint of a sharp superconducting transition occurs at ~ 4.6 K. Due to the sample morphology, it is impossible to measure directly the inter-chain resistivity along the other two orthogonal directions, ρ_{c^*} and ρ_{a^*} . Instead, we use the anisotropy in its upper critical field to evaluate its resistivity anisotropy. According to anisotropic Ginzburg-Landau theory, the upper critical field H_{c2} with a field applied along i direction is $H_{c2}^i = \frac{\Phi_0}{2\pi\xi_j\xi_k}$, where Φ_0 is the fluxoid and $\xi_{j,k}$ ($\propto v_{j,k}$) is the coherence length in the directions orthogonal to the field. On the other hand, the resistivity ρ_i is inversely dependent on the square of the Fermi velocity, $\rho_i \propto \frac{1}{v_i^2}$. These combined give the following relations: $\frac{H_{c2}^i}{H_{c2}^j} = \frac{\xi_i}{\xi_j} = \frac{\sqrt{\rho_j}}{\sqrt{\rho_i}}$. Fig. 2 displays the temperature dependence of the b -axis resistivity under several values of magnetic field applied along the three orthogonal directions, along with the

cumulative phase diagram of H_{c2} , using the criteria of the midpoint of the transition. The resultant H_{c2} extrapolated to $T=0$ K is 5.4 T, 9.4 T and 3.3 T, for $H \parallel a^*$, $H \parallel b$, $H \parallel c^*$, respectively, in agreement with previous measurements[25, 29]. Therefore, the resistivity anisotropy $\rho_{a^*}:\rho_b:\rho_{c^*}$ is estimated to be 3:1:8. This anisotropy is rather small compared with other Q1D materials, like (TMTSF) $_2X$ ($X=PF_6$, ClO_4), $Li_{0.9}Mo_6O_{17}$ and $PrBa_2Cu_4O_8$.

Fig. 3 shows a series of field sweeps at fixed temperatures in the normal state of one sample studied (hereafter labelled as #1). In $H \parallel a^*$ and $H \parallel b$ configurations, the field is swept up to 9 T, while for $H \parallel c^*$, the maximum field is 13 T. It is evident that the MR is nonquadratic and can be best fitted to a power-law dependence, $\Delta\rho/\rho \propto B^\alpha$. While the exponent α shows a slight increase with the increasing temperatures, it is close to 1 in the whole temperature range studied. The magnitude of the MR is comparable when field is aligned along the a^* - and c^* -axes, and is the smallest with field pointing to the chain direction. The linearity of the MR is optimal at temperatures near 30 K. It is worth noting that the quasi-linear MR is not likely derived from the superconducting fluctuations to such high temperatures, as even in the under-doped high T_c cuprates with strong superconducting fluctuations, the fluctuations extend only to a temperature no more than 5 times of T_c [30, 31].

In order to see if this power-law like MR will ever saturate at a higher field, we study the transverse MR ($H \parallel c^*$) of a second sample (#2) in the pulsed magnetic field. As seen from Fig. 4, this quasi-linear MR shows no sign of saturation up to 50 T. In general, the transverse MR of a metal will saturate in the high fields once $\omega_c\tau \gg 1$ unless the material is perfectly compensated or it has open orbit in the FS[11, 32]. In $Ta_4Pd_3Te_{16}$, according to band structure calculations[28], its FS contains a 2D hole cylinder ' α ', two nested 1D sheets ' β ' and ' γ ', and a 3D sheet ' δ '. Hence, the open orbit associated with the 1D Fermi sheets may be responsible for the nonsaturating MR observed here.

Fig. 5 collectively shows the exponent α in B^α dependence of the MR at different temperatures and field configurations from four samples studied (the other two labelled as #3 and #4). There are total 48 points in this figure. The most striking feature of this figure is that, most of the data points (especially below 50 K) resides in the range of $\frac{4}{5}$ to $\frac{4}{3}$, although α increases slightly with temperature.

In recent years, linear MR has been widely observed in the Dirac materials, such as topological insulators[12], 3D bulk materials with Dirac states[13], and iron-based superconductor $BaFe_2As_2$ [14]. The linear energy dispersion of Dirac fermions leads to nonsaturated linear MR once the field exceeds a critical field B^* such that all states occupy the lowest Landau level in the quantum limit. Therefore, the transverse MR displays a crossover from the low- B quadratic dependence to the high- B linear MR at the critical field B^* . The crossover field B^* has the T -dependence: $B^* = \frac{1}{2e\hbar v_F^2}(E_F + k_B T)^2$, where v_F and E_F are Fermi velocity and Fermi energy respectively[13]. In $Ta_4Pd_3Te_{16}$, however, its low- B MR profile can not be fitted to the quadratic form in a reasonable field window. In addition, the above T -dependence of the critical field is not observed either.

In silver chalcogenides, $Ag_{2+\delta}Se$ etc., the resistance exhibits an unusually linear dependence on magnetic field without any signs of saturation at fields as high as 60 T[15, 33]. The underlying physical origin of the linear MR in this non-magnetic material remains controversial[34, 35, 36]. A plausible explanation is the conductivity fluctuations associated with inhomogeneous distribution of silver ions[35]. In our $Ta_4Pd_3Te_{16}$ crystals, the sample is in single phase and the quality is high, confirmed from both XRD and energy dispersive x-ray spectra, hence this mechanism is unlikely here.

As described earlier, the linear positive MR has also been observed in some ferromagnets, such as the cobalt-doped FeSi[15]. This linear MR was attributed to the quantum $e-e$ interference interaction. The effect of a magnetic field on the $e-e$ interaction was derived three decades ago in the nonmagnetic cases[37]. Under this circumstance, the magnetic field induces a spin gap ($\propto g\mu_B H$, where g is the Lande factor and μ_B is the Bohr magneton) which suppresses the contribution of $e-e$ interaction to the conductivity and leads to a positive MR proportional to $\ln(g\mu_B H/k_B T)$ in 2D and to $\sqrt{g\mu_B H}$ in 3D[37]. In a material with ferromagnetic correlations, however, there exists an (exchange) gap in the absence of the external field. The external field further increases the gap and induces a correction to the resistivity which is linearly proportional to H at any laboratory field. However, there is no observed experimental evidence to date in favor of such ferromagnetic correlations in $Ta_4Pd_3Te_{16}$. Instead, both the pressure and STM study suggested that the material may be actually close to a CDW instability[25, 27]. The question therefore remains of how the electrons scatter off the CDW fluctuations and ultimately lead to a linear MR seen in this study.

At last, let us examine the Kohler's rule in this Q1D material. Kohler's plot, as exemplified for sample #1, is shown in Fig. 6(a)-(c) for three different field orientations respectively. Clearly, the Kohler's rule is violated, in particular below ~ 50 K, in all field directions studied here. Generally, the departure of the Kohler's scaling results from the loss of the carriers or the anisotropic scattering $\tau(k)$ that does not have the same T -scaling on different portions of the FS. Prior to the report of Ref. [22], Kohler's rule was widely believed to be violated in cuprate superconductors as a result from the two-lifetime scattering and non-Fermi liquid excitations. In the disordered Q1D $PrBa_2Cu_4O_8$, Kohler's rule was seen to be violated whereas it was obeyed in the pure, clean samples. This dichotomy was proposed to arise from the disorder-tuned dimensional crossover from 3D to pure 1D and the corresponding spin-charge separation in the 1D TLL regime. Given the resistivity anisotropy quoted above, it is farfetched to assign $Ta_4Pd_3Te_{16}$ into a TLL material. Instead, it is natural to deem that, as inspired by the pressure study[25], the material is on the border of a CDW instability and the violation of Kohler's rule is the result from the gapping out of the electrons with decreasing T due to the density-wave formation[27].

4. Conclusion

In conclusion, we have uncovered an anomalously quasi-linear MR, irrespective of the field directions, in the normal state of the Q1D $Ta_4Pd_3Te_{16}$ superconductor. This quasi-linear MR shows nonsaturating behavior up to 50 T, the highest field in this study. Moreover, the Kohler's rule was seen to be violated in all three field directions studied. In combination with the previous report of its T_c -pressure diagram, it is tempting to link our observation to the proximity to a CDW instability. In this respect, it is interesting to see how the Drude tail in the optical response evolves in the T range studied here. The origin of the quasi-linear MR observed in this study however invokes more theoretical investigations in the future.

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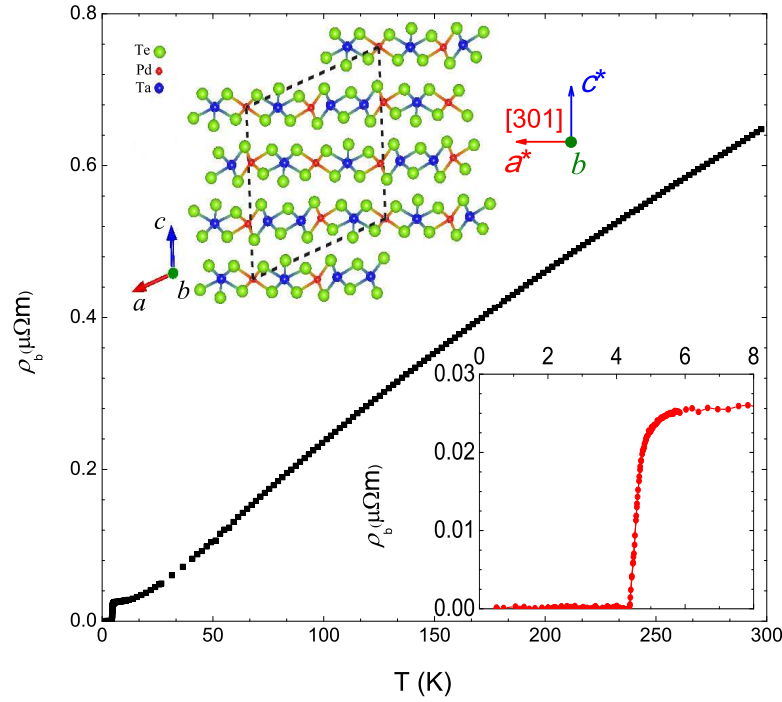


Figure 1. (Color online) The representative zero-field resistivity as a function of temperature, with the bottom-right inset blowing up the superconducting transition. Upper left inset: Schematic representation of the crystallographic structure of $Ta_4Pd_3Te_{16}$ as seen from a perspective along the b -axis. Pd atoms (in red) are octahedrally coordinated by Te (in green), forming edge-sharing $PdTe_2$ chains along the b axis. Te atoms display both prismatic coordination and octahedral coordination around the Ta sites (in blue), forming $TaTe_3$ chains and Ta_2Te_4 double chains. A new a^* -axis is defined in flat Ta-Pd-Te layers normal to the b -axis, as seen from the inset and the c^* -axis is perpendicular to the a^* -axis. The unit cell is depicted by the thin dashed line.

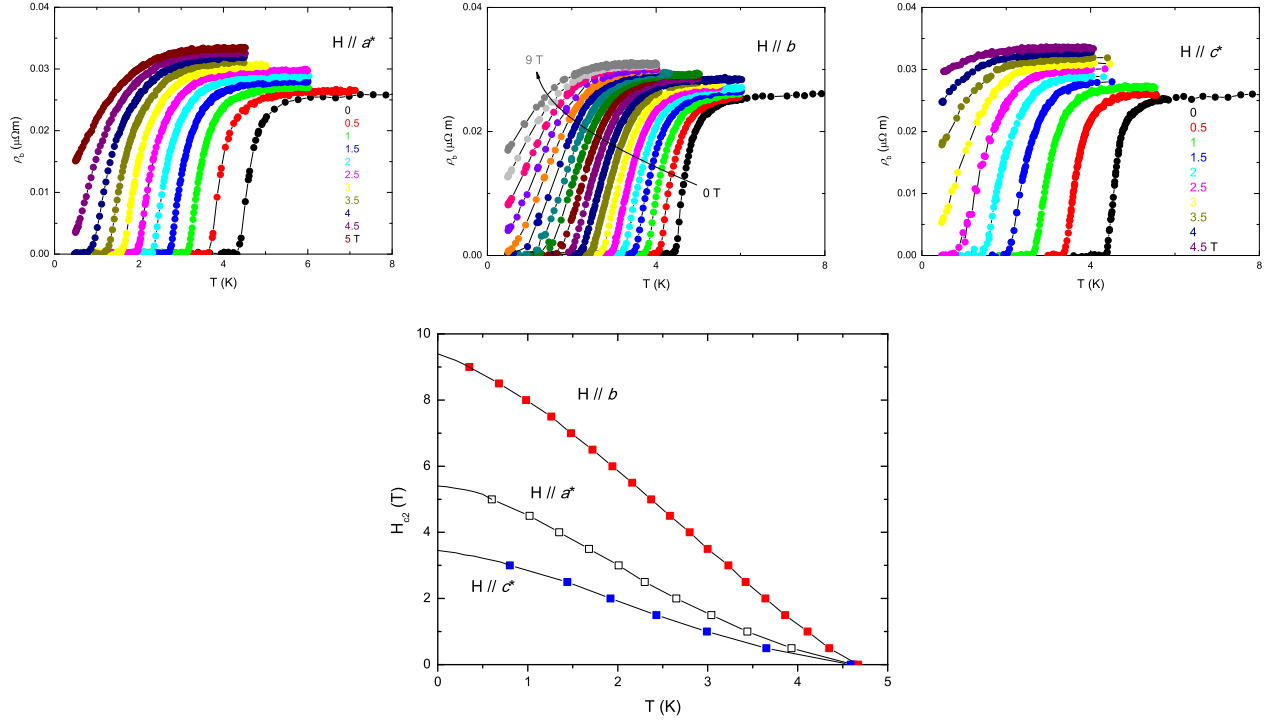


Figure 2. (Color online) The top three panels show the T sweeps of the b -axis resistivity of $\text{Ta}_4\text{Pd}_3\text{Te}_{16}$ under various fields for H aligned along the three orthogonal axes. Bottom panel: H - T phase diagram using the midpoint of the transition for field aligned along the a^* , b , and c^* directions.

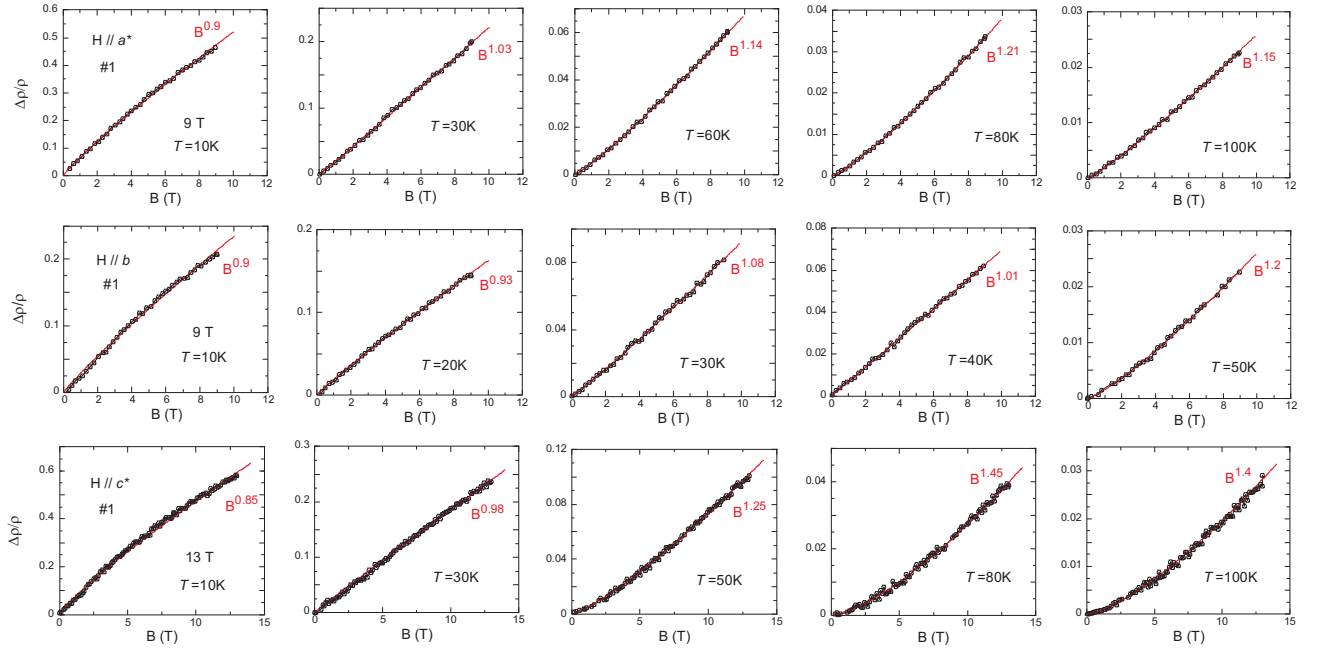


Figure 3. (Color online) The field sweeps at several constant temperatures for $H \parallel a^*$, $H \parallel b$ and $H \parallel c^*$, respectively, for sample #1. For the former two field directions, the field is up to 9 T while for the latter, is up to 13 T. The data are fitted to B^α , with α given in the individual figures.

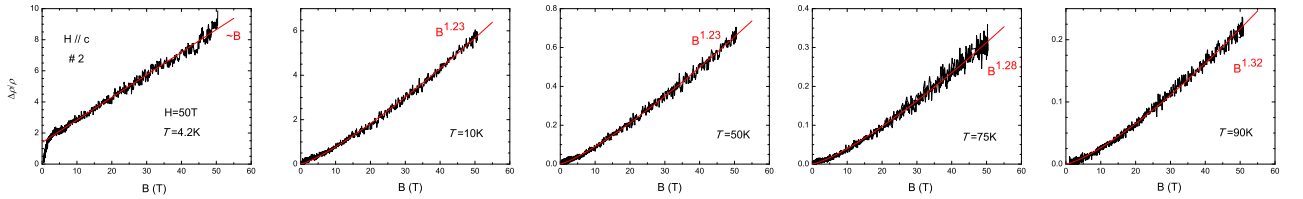


Figure 4. (Color online) The B -sweeps at constant temperatures in the pulsed magnetic field along the c^* -axis up to 50 T. The data are fitted to B^α , with α indicated in each panel. The drop at low B for $T=4.2$ K panel is due to superconductivity at this temperature.

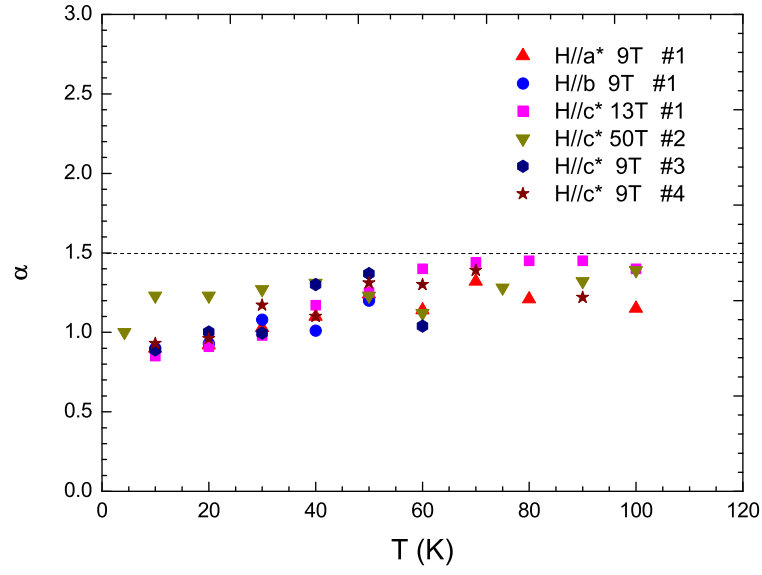


Figure 5. (Color online) The exponent α in the power-law fitting B^α , collected from 4 different samples under three different magnets and field directions. Total 48 points are given in the plot.

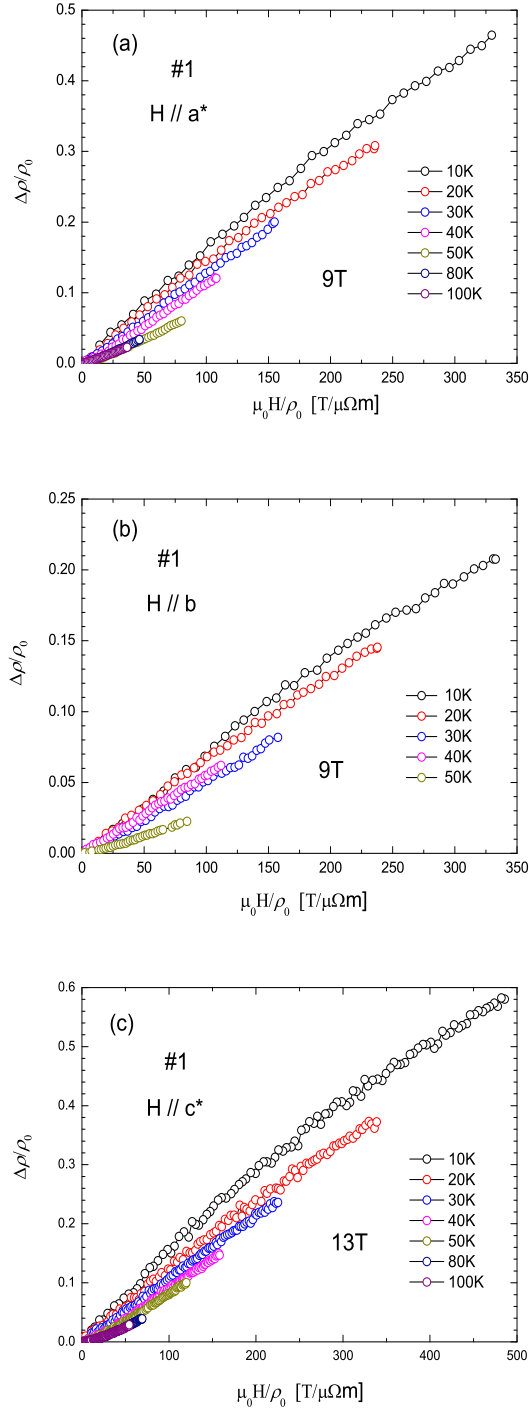


Figure 6. (Color online) Panels (a)-(c) show the Kohler's plots for sample #1 with $H \parallel a^*$, $H \parallel b$ and $H \parallel c^*$, respectively. For $H \parallel a^*$, $H \parallel b$, the maximum field is 9 T and for $H \parallel c^*$, maximum field is 13 T.